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REVIEW OF WATER MONITORING PROCEDURES AT CLINTON LABORATORIES

by

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REVIEW OF WATER MONITORING PROCEDURES AT CLINTON LABORATORIES

Introduction

The methods used for water monitoring were set up at a time when it was anticipated that the water activity would be low at all times. Therefore, GM counter methods were used to obtain sensitivity at the expense of accurate interpretation of the results. It was expected that sufficiently accurate information for extrapolation to the W levels would be obtained and that no significant levels for health hazard at Clinton would arise.

It was further assumed from the alleged hazards of ingestion of the several fission products, and from the estimated composition of the mixture in the overflow water that the limiting hazard would be that of external radiation to a body immersed in the water, rather than that of ingestion.

The tolerance dose for an immersed body has been set at 100 mrep/24 hours.* This refers then to the extremely improbable circumstance of continuous immersion of the body in water. Alternatively it is supposed that the radiosensitivity of a fish, at all stages, is similar to that of a human being. For ingestion, the human was supposed to take 3 liters of water per day. The absorption process in a fish was not considered well enough known.

Measurement of β ray dose.

The rate of energy absorption in tissue or water substance corresponding to a dosage rate of 100 mrep in 24 hours is 0.98×10^{-11} watt/gm, or sufficiently closely 10^{-11} watt/gm. If the water contains an active substance giving electrons of average energy E Mev, the tolerance concentration is

$$\frac{1.6}{E} \ \mu c/\text{liter}.$$

Suppose the activity of the water is due to UX_2, which gives electrons of energy 2.32 Mev with average energy E about 1.0 Mev. The tolerance concentration will be 1.6 μc/liter. This is equivalent to 3550 counts/min/ml.

Experimental Standard

A "tolerance water" was prepared experimentally in the following manner:

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* 1 rep (roentgen-equivalent physical) is the quantity of radiation that gives rise to an energy absorption of 83 ergs/gram of tissue. For quantum radiation 1 rep = 1 r.

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A weighed quantity of UNH was dissolved in water and a thin-walled β -ray chamber in a lusteroid tube was suspended in it. The observed dosage-rate was 70 mrep/hr. The lusteroid wall weighed 46.1 mg/cm^2, and the chamber wall 7.3 mg/cm^2.

It was assumed that the mass absorption coefficient was 6. Hence the absorption correction factor should be

$$e^{6 \times 0.0535} = \underline{1.38}$$

and the true dosage-rate 97 mrep/hour. Tolerance water was made by dilution of the solution.

This "tolerance water" was evaporated in 5 cc. samples on to porcelain dishes and used as the standard for β -counting of other samples. These counts have been made by G. W. Struthers who found 470 counts/min/ml at 10% geometry. Under these conditions about 30% of the count is due to scattered electrons. The true count is therefore

$$\frac{470 \times 10}{1.3} \text{ c/min/ml} = 3620 \text{ counts/min/ml.}$$

This seemed to be in good agreement with the expected count. However, it now seems that such an agreement was largely fortuitous and that it postponed a more careful consideration of the method.

Corrections to the "tolerance water"

1. Specific Gravity

The strong solution used to give readings with the small chamber in a reasonable time had a specific gravity of 1.4. The effective radius from which electrons could enter the chamber was therefore less than the range in water and the measured dosage-rate lower. When the solution was diluted the specific gravity became approximately 1. Estimated dosage-rate of the tolerance water was probably too high by a factor of $\sim$ 1.4. Correction factor = $\underline{0.72}$.

2. Filtration

For simplicity, the absorption in the lusteroid wall was considered only in a plane perpendicular to the axis of the cylindrical chamber. The functions governing absorption in a cylindrical wall have been partially evaluated by R. R. Coveyou, but there is no available solution for this case. If the radiation had been absorbed through a large flat sheet, it would have been governed by an F_1 function, and the absorption correction would be 2.0 (see CH-930). The average of 2.0 and 1.38 would be a reasonable value. Correction factor = $\frac{1.38}{1.7} = \underline{0.81}$.

3. Effect of Evaporation

UNH gives, in addition to the UX_2 electrons of energy 2.32 Mev. an equal number of electrons of energy 0.15 Mev from UX1. In the liquid the effect of the latter would be unimportant, giving only 1/16 of the total energy. Moreover, in tissue, the 10 mg/cm² layer of passive absorption would absorb a large fraction of these rays. In the experimental set-up none could penetrate the lusteroid tube.

When the active water was evaporated to dryness, the UX_1 and UX_2 electrons counted equally in the absence of filtration. Approximate calculations from the thickness of the sample, the air absorption, and the counter window of 4 mg/cm² suggest that about 20% of the UX_1 particles would be counted. Correction factor $= \frac{1}{1.2} = 0.83$

The electron count due to a tolerance water at 10% geometry becomes 470 x 0.72 x 0.81 x 0.83 c/min/ml = 227 c/min/ml

This is equivalent to $\frac{2270}{1.3} = \underline{1750}$ true counts/min/ml.

This is approximately one-half of the theoretical value.

4. Effect of energy on the measurements

When a proper sample has been set up, it is a straight forward procedure to test any other sample giving disintegrations of the same average energy. For other energies the permissible concentration will be inversely proportional to E, but the counter procedure will give only values corresponding to the curie content. It has been considered standard practice to recheck any strong samples by an ionization method. Without such a check, the standardization by UNH will give conservative results for all reasonable fission product mixtures. For a fission mixture a sample that gives 227 c/min/ml probably gives no more than 50 mrep/day of β radiation.

5. Geometrical factor

The existing standardization procedure was based on the energy absorbed per gram of tissue from radiation incident in all directions. In all practical cases* the β-radiation is incident from a hemisphere only. The permissible count would then be doubled, namely 454 c/min/ml. This is so close to the current 470 c/min/ml that no change is recommended until the whole sampling procedure is revised.

The β-radiation from water giving B c/min/ml under the present procedure is then

$$\frac{100 \times B}{24 \times 470} \text{ mrep/hr} = \underline{0.0088} \times B \text{ mrep/hr}$$

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*One might picture that an ear could receive radiation both fore and aft. Fish eggs, etc. would receive radiation from a full sphere.

Measurement of gamma-ray dose

The gamma ray dosage rate at the center of a sphere of active liquid of radius R is

$$G\,(1 - e^{-\mu R})$$

where G = dosage-rate at center of an infinite sphere and μ = linear absorption coefficient of the radiation in water.

μ for hard radiation is $\sim 0.04\ \mathrm{cm}^{-1}$. Therefore a sphere of radius 17 cm will give half the dosage-rate of an infinite sphere. This gives a basis for reasonable sample sizes for gamma activity monitoring.

(1) Immerse the measuring device at the center of a vessel of 18" radius. A single reading cannot be in error by more than 20%, due to finite sample size. The method requires a 100 gallon sample and is only practicable when the measuring equipment can be taken to the active water.

(2) Immerse the measuring device successively at the center of two vessels of radius R and 2R where R is about 3-1/2".

If S = dosage rate in small vessel
L = dosage rate in large vessel

$$S = G(1 - e^{-\mu R})$$

$$L = G(1 - e^{-2\mu R})$$

$$S^2 = G^2(1 - 2e^{-\mu R} + e^{-2\mu R})$$

$$(2S-L) = G\,(1 - 2e^{-\mu R} + e^{-2\mu R})$$

$$\therefore G = \frac{S^2}{(2S-L)}$$

This extrapolation to the dosage-rate at infinite volume can be used with ion chamber measurements or with counters. There are some objections -

(1) The difference term (2S-L) leads to inaccurate results unless S and L are well determined. A 10% error in S or L can give 50% error in G.

(2) The formula is valid for a single active material of absorption coefficient μ. The influence of mixed activities is less than expected. It cannot lead to an error of more than 20%.

(3) With counters, there is an unknown factor of wavelength dependence, which is especially complicated by the degradation of radiation within the liquid. Brass counters were used initially. More recently it has been necessary to resort to glass counters with silvered walls. Both are poor from the wavelength dependence point of view.

(4) The finite size of the measuring device perturbs the above equations. The effect is small if the diameter of the chamber or tube is less than 1"

(5) Suspended matter in the active water settles out during the sample process. This spoils the present readings and leaves the equipment badly contaminated.

Nevertheless the gamma sampling method is useful whenever it is inconvenient to take measuring equipment to the water supply. Results obtained to date have checked rather well with direct ionization measurements in the ponds.

The brass counters used gave 3350 c/min when exposed to 1 mr/hr of Ra γ radiation. Gamma-ray dosage rate is thus -

$$\frac{S^2}{(2S-L) \times 3350} = \frac{3 \times 10^{-4} \times S^2}{(2S-L)} \text{ mr/hr}$$ where S and L are expressed as C/min from the small and large samples respectively.

Relation of β and γ activity

It was originally hoped that the ratio of γ to β activity would be approximately constant. The ratio has ranged from 0.4 to 4.0, which is well beyond the possible errors of both systems. Hence it is necessary either to use a measuring scheme that records the combined β and γ dosage, or to observe each separately. The latter is the preferred method despite the additional equipment requirements.

Hazard of ingestion relative to external radiation

The assumption that the hazard from 24 hour immersion exceeded the hazard from ingestion of normal quantities of the water was apparently well-founded in the case of the Clinton overflow. Nevertheless the ingestion hazard is such a rapidly varying function of the composition of the active materials that it will never be safe to proceed without analyses for some special elements.

For the purposes of this review, tolerance concentrations of fission products given in CL-697, Chap. 12 Sec. A and C will be accepted. These are less conservative than figures given by J. G. Hamilton by a factor of about 100. The higher figures are based on a permissible radiation of 100 mrep/day and an intake of 3 liters of water; the lower figures on 10 mrep/day and 10 liters intake. These differences emphasize the present large uncertainties in assessing the ingestion hazards. Only the case of repeated intake from a contaminated water source of maintained concentration need be considered. The ingestion hazard resolves into two components -

(1) effect on bone marrow
(2) effect on gut

If it can be assumed that there is no abnormal hold up of active material in the gut, and that the radiation sensitivity is essentially the same as that of the skin and superficial tissues, then the effect on gut can be ignored. The gut dose will necessarily be less than the skin dose for 24 hour immersion. Elements notable proceeding to the bone are -

	Tolerance Concentration	
	Bone Marrow	Skin
Ba	3.1 μc/liter	3.2 μc/liter
Sr	0.07	1.9
Ru	3.4	11.0
Mixed FP	1.2	$\sim$ 3

Whether the limiting hazard is the radiation to bone marrow or to skin is controlled almost entirely by the content of active strontium. If more than 1% of the total activity is due to strontium the tolerance concentration cannot be determined without an analysis of the water.

The waste waters from the plant have been diluted by raw river water which has fortunately precipitated most of the barium and strontium. At the present time the precipitation is encouraged by the addition of calcium salts. An analysis of the W6 overflow water by J. G. Hamilton and collaborators indicates that the filtrate activity is made up of 0.27% (Ba + Sr), 58.5% Zr, 25.6% Cb, 1.69% Ru and less important FP's. A sediment in the waste has 3.79% (Ba + Sr) but it contributes only 2.4% to the total liquor activity.

The tolerance concentration for ingestion of the filtrate is 21.6 μc/liter. For external radiation it is 8.6 μc/liter.

The ingestion tolerance concentration for the precipitate is 1.7 μc/liter, and for the total liquor is 16.8 μc/liter.

As far as external radiation is concerned the liquor presents a greater hazard from γ radiation than from β. The tolerance concentration for the γ radiation alone is 2.3 μc/liter. For the combined radiations it is 1.8 μc/liter. Therefore, at the present time, the hazard of ingestion is not the controlling feature.

Disposition of activity after release from the holdup ponds.

There is no problem attached to the maintenance of water activity at levels below 100 mrep/day at the pond discharge. This corresponds to 1 to 2 μc/liter or, for a daily discharge of 600,000 gals, a daily discharge of 2-1/2 to 5 curies. On the average, no more than this can proceed through the dam per day. Since the average flow of the Clinch River is 4000 c ft/sec, the average concentration in the river will be 10^{-4} μc/liter. At minimum flow the concentration would be 5 x 10^{-2} μc/liter.

In fact, relatively little of the activity goes through the dam. It is well known that the active materials are readily absorbed in the clay in the White Oak Creek and in the dam. During a two month period in which approximately 5 curies of active material were discharged per day, the activity of the clay in the creek rose to 4 μ c/gm of dried clay. For a long period the value might rise to 30 μ c/gm. In the dam itself the activity would probably not exceed 0.1 μ c/gm. The expected hazard from the mud activity would be 4-fold.

(1) Beta and gamma radiation from the source

(2) Absorption of activity by plants and subsequent ingestion of the plants by animals or humans.

(3) Absorption by fish, especially scavengers

(4) Discharge to the river by flood.

(1) and (2) can be controlled by suitable fencing of the creek and dam, (3) is governed by fish screen at the dam, (4) is rather an unknown quantity. The equilibrium amount of activity in the dam system would be between 1000 and 2000 curie. If all this activity were discharged at once, either the Clinch River would be naturally at flood levels or could be made so by the TVA system. At least 50,000 c.ft/sec would be available. This would give a concentration of about 10^{-2} μ c/liter. Moreover, if all the activity accumulated at one place in the river bed it would not present a hazard in a river 6 feet deep. The risk of ingestion by a fish and subsequent consumption as food would be real. But, in the event of a catastrophe of the postulated nature, severe control measures would not be out of line.

A permissible discharge of 5 curies per day of waste water of <u>approximately the present composition</u> is less conservative than that given by J. G. Hamilton (letter to S. T. Cantril 6/20/44). The latter was inadvertently based on the <u>minimum</u> Clinch flow. It would probably be agreed that the actual limiting hazard is gamma radiation from the creek or from the dam pond at low water. Permission to fence off this area as widely as necessary, together with adequate patrol would be sufficient protection.

The maximum waste discharge is therefore governed largely by security, since an elaborate fence and patrol system outside the plant site might excite interest. It might be possible to add some noxious agent to the water to discourage drinking by pastured animals and swimming. The concentration would require to be such that dilution by the Clinch would leave the water palatable.

<u>Preliminary suggestions for revised procedures.</u>

Improved design of equipment is currently being considered by J. H. Hall. General principles only are discussed in this review.

β ray dosage

(a) Ion chambers are preferable to counters because the ionization function more closely follows the biological hazard. A thin window Lauritsen electroscope would be adequate. Window should be 10 mg/cm^2 to correspond with layer of passive absorption. There is no further interest in quantitative measurements on very weak samples, where the electroscope technique would be laboriously slow.

(b) The water should be sampled without evaporation to prevent emphasis of a low energy component. Either a chamber immersed in the liquid or immediately over a liquid layer of depth about 1 cm should be used. The risk of spilling and contamination is large. The equipment should be in a special water sampling room.

(c) For continuous monitoring of overflow the water should flow over a shallow tray with the chamber above it. The apparatus has to be shielded from gamma rays from the main body of water.

Gamma ray dosage

(a) Ion chambers are preferable to counters by virtue of superior wave-length dependence. Very weak samples can be tested qualitatively by a counter method. The sampling technique in buckets of radii 3-1/2" x 7" is acceptable.

(b) When equipment can be taken to the disposal basin, the following procedures can be used -

1. Sample at the middle of lead protected buckets of radii 3-1/2" and 7". Either two chambers or two counters can be arranged to register simultaneously or alternately (to save equipment) on a ~~recorder~~ meter. A conversion chart for translating all combinations of S and L can be written.

2. Sample with a single chamber or counter in a region surrounded on all sides by at least 18" of water. The principal trouble in this technique is that activity tends to collect in the mud at the base of a pond or to float in a scum on the surface. Erroneous results can then arise, fortunately in the direction of over-caution.

3. Float a chamber or counter on the surface and read one-half the dosage for full immersion. This is valuable in a rather shallow pond, where immersion brings the meter to a point where it records mud activity. Equipment can be kept free from contamination by this method. Moreover a single chamber with a rotating shutter could be set to record γ radiation and ($\beta + \gamma$) radiation in such a manner that the two could be separated and corrected for different geometry.

<u>General</u>

1. An analysis for the active materials with special reference to the percentage of Ba, Sr and Ru should be made at intervals to ensure that the hazard of external radiation is in fact the controlling feature.

2. The deposition of activity on mud etc., should be checked. Human and animal approach to active areas should be restricted. The lower gate of the dam should always be closed.

3. The [illegible]rds of activated fish should be considered.

4. The presence of alpha ray emitters of long life should be checked.

5. For systems other than that at Clinton the same procedures apply with individual consideration of the disposal of water beyond the point of control.